

# Ground-based FTIR measurements of O<sub>3</sub>- and climate-related gases in the free troposphere and lower stratosphere.

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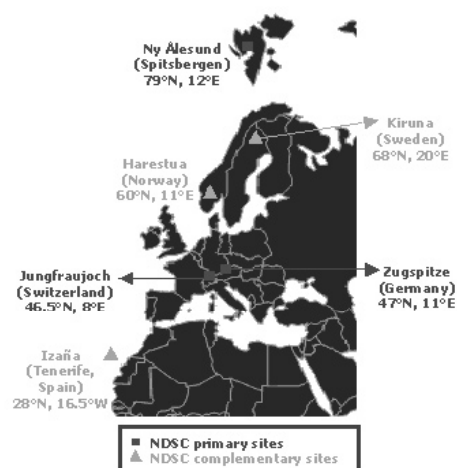
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**Abstract.** In the frame of the EC project UFTIR (Time series of Upper Free Troposphere observations from a European ground-based FTIR network), a common strategy for an optimal determination of the chemical composition in the free troposphere and lower stratosphere with ground-based Fourier-transform infrared (FTIR) spectrometers is being developed. The project focuses on 6 target species that are O<sub>3</sub>, CO, CH<sub>4</sub>, N<sub>2</sub>O, C<sub>2</sub>H<sub>6</sub> and CHClF<sub>2</sub> (HCFC-22). The strategy consists in selecting the most appropriate parameters to retrieve vertical concentration profiles from solar FTIR spectra. Among the important parameters are the spectral microwindows: they have been optimised to maximise the information content and to minimize the influence of poorly known spectroscopic data and interfering species.

## Introduction

The UFTIR network (Figure 1) includes six stations in Europe that contribute to the Network for Detection of Stratospheric Change (NDSC) with operational high-resolution FTIR spectrometers since at least a decade. Time series of total column abundances for a large number of atmospheric gases are archived at the NDSC database (<ftp://ndsc.wv.noaa.gov>). In addition for a number of trace gases the concentration profiles up to approximately 35 km can be retrieved using recently developed inversion algorithms, like SFIT2 and PROFITT (Hase et al., 2004), by

analysing the shapes of infrared absorption lines. In UFTIR, the available time series of spectral data at each station will be re-analyzed to derive the volume mixing ratio profiles for the target gases ozone (O<sub>3</sub>), carbon monoxide (CO), methane (CH<sub>4</sub>), nitrous oxide (N<sub>2</sub>O), ethane (C<sub>2</sub>H<sub>6</sub>), and CHClF<sub>2</sub> (HCFC-22).



**Figure 1.** The UFTIR network spanning 28°N to 80°N.

A prerequisite for this analysis is to select optimum microwindows and spectroscopic parameters. This selection will determine to a large extent the FTIR inversion

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capabilities to deliver, e.g., free tropospheric and lower stratospheric abundances of the target species. This paper discusses the optimal retrieval options per target molecule that will be adopted (with possibly minor modifications) in the UFTIR time series analyses.

## Retrieval strategies and capabilities: overview

Table I summarizes the main results as to retrieval strategy and performances for the UFTIR molecules. Generally HITRAN2000 and its official updates have been adopted as the spectroscopic database, except where indicated otherwise. The next sections discuss retrieval particularities for each UFTIR species.

**Table I.** Summary of best retrieval microwindows and performances. DoFs indicates the approximate number of independent elements of altitude-resolved information calculated for a solar zenith angle around 60°; the precise value depends on the particular retrieval conditions like spectral signal-to-noise ratio, a priori information, station altitude, etc. Alt<sub>max</sub> indicates the approximate highest altitude for vertical inversion.

	Microwindow (s) (cm <sup>-1</sup> ) used simultaneously	Interfering species;	DoFs / Alt <sub>max</sub> (km)
O <sub>3</sub>	1000.0 – 1005.0 1110.0 – 1113.0 1117.3 – 1117.9 1120.1 – 1122.0	H <sub>2</sub> O, N <sub>2</sub> O, CH <sub>4</sub>	5 / 35
CO	2057.70 – 2057.91 2069.55 – 2069.72 2157.40 – 2159.20	H <sub>2</sub> O, N <sub>2</sub> O, O <sub>3</sub> , solar lines	4 / 18
C <sub>2</sub> H <sub>6</sub>	2976.50 – 2977.20	H <sub>2</sub> O, O <sub>3</sub> , CH <sub>4</sub>	2 / 30
N <sub>2</sub> O			
Option 4 μm	2481.30 – 2482.60 2526.40 – 2528.20 2537.85 – 2538.80 2540.10 – 2540.70	H <sub>2</sub> O/HDO, CO <sub>2</sub> , O <sub>3</sub> , CH <sub>4</sub>	4.5 / 30
Option 8.5 μm	1161.34 – 1161.66 1182.49 – 1182.83 1183.25 – 1183.74	O <sub>3</sub> , HDO, CH <sub>4</sub>	4.5 / 30
CHClF <sub>2</sub> (HCFC- 22)	828.8 – 829.35	H <sub>2</sub> O/HDO, CO <sub>2</sub> , O <sub>3</sub> , ClO, C <sub>2</sub> H <sub>6</sub> , solar lines	1 / 25

## Ozone (O<sub>3</sub>) retrieval

O<sub>3</sub> will be retrieved essentially in the 1000-1005 cm<sup>-1</sup> window. This window contains a large number of O<sub>3</sub> absorptions of different strengths and temperature dependencies, thereby increasing the vertical information content (Barret et al., 2002). The main interfering absorber is H<sub>2</sub>O. Because of its high variability, the best approach to deal with it is to first retrieve the H<sub>2</sub>O profile in the three nearby windows (1110-1120 cm<sup>-1</sup>) and to use this result as a priori in the fit of all 4 windows simultaneously.

## Carbon monoxide (CO) retrieval

Under humid conditions (e.g., tropical low altitude station) it may be advantageous to first retrieve the H<sub>2</sub>O profiles and to use these results as a priori in the CO retrieval.

## Ethane (C<sub>2</sub>H<sub>6</sub>)

The 2976.61-2976.66 cm<sup>-1</sup> range in the selected micro-window for C<sub>2</sub>H<sub>6</sub> contains an unknown feature - it is therefore 'deweighted' in the retrieval. The spectroscopy to

be used for the <sup>P</sup>Q<sub>3</sub> branch of C<sub>2</sub>H<sub>6</sub> is the one from Pine and Rinsland (1999); the spectroscopic parameters of the O<sub>3</sub> triplet at 2977 cm<sup>-1</sup> have been revised for the purpose of UFTIR (Mikhailenko et al., 2002). As for ozone, the H<sub>2</sub>O profile must be retrieved in a first step and this retrieval result must be used when fitting the C<sub>2</sub>H<sub>6</sub> profile.

## Nitrous oxide (N<sub>2</sub>O)

N<sub>2</sub>O can be retrieved around 4 μm as well as around 8.5 μm. Retrieval tests on both spectral regions indicate good agreement of the retrieval results, and a bias of order 2% between the total column values (2% higher for option 4 μm). Analyses of time series will confirm the best selection and associated error budgets.

## CHClF<sub>2</sub> (HCFC-22) retrieval

The retrieval of HCFC-22 requires initial retrievals of the H<sub>2</sub>O and then the O<sub>3</sub> profiles, and using these results, with scaling, in the HCFC-22 retrieval. The other interfering gases can be dealt with by scaling only. The cross-section line of HCFC-22 must be shifted by +0.0035 cm<sup>-1</sup>.

## Methane (CH<sub>4</sub>) retrieval

CH<sub>4</sub> has not been included yet in Table I because it was found that uncertainties in the actual knowledge of its spectroscopic parameters are too large to obtain reliable retrievals. Laboratory work is being undertaken in the frame of the project to reduce these uncertainties.

## Synthesis

A revised choice of spectral microwindows, associated spectroscopic data and retrieval approaches, has improved the retrieval characteristics for most of the UFTIR target species. The present choices will be applied systematically to the re-analysis of available time series at all stations involved. The focus will then shift to the optimisation of additional retrieval parameters like the a priori information. The project partners have established firm links with the international NDSC (Network for the Detection of Stratospheric Change) FTIR community, thus supporting the implementation of the developed strategy on a quasi-global scale and providing consistent support for model developments and for validation of satellite experiments.

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